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Temperature distribution and modification mechanism inside glass with heat accumulation during 250 kHz irradiation of femtosecond laser pulses

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Heat accumulation by high repetition rate femtosecond laser irradiation inside glass generates a much larger modification than that by a single pulse. In this study, we determined the temperature distribution due to heat accumulation and the characteristic temperature for heat modification inside a soda lime glass by analyzing the relationship between the radius of modification and glass temperature. The validity of the analysis was confirmed by reproducing the modification due to two-beam irradiation. The determined characteristic temperature suggested that the temperature distribution and the spatial dependence of the stress relaxation are important in the mechanism of heat modification. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3046101]

When femtosecond laser pulses are focused tightly inside a glass, the material in the laser focal region is modified as a result of local nonlinear photoionization.[1–11] Because a large amount of light energy is absorbed by electrons and the energy is transferred to the lattice,[12] the temperature around the photoexcited region increases to as high as several thousand °C.[13,14] When only one laser pulse is focused by a microscope objective, the temperature increase in the surrounding material is too small to induce structural change in the surrounding region because of a small photoexcited volume (for example, 10 μm^3 with 20× objective lens). On the other hand, when many femtosecond pulses are focused at a high repetition rate (more than several hundred kilohertz), the heat accumulation occurs and results in modification in the surrounding material.[3–6] Recently, the heat accumulation in femtosecond laser machining has attracted interest because this effect is important in the fabrication of optical waveguides[3,4] and in inducing crystallization[8] or ion migration around the laser focal volume inside the glass.[9,10]

The morphology of the modification in a glass due to heat accumulation has several circular structures, as shown in Fig. 1. The inner structure should be attributed to the photoexcitation and modification due to shock wave generation.[13] The outer structure has been attributed to be the result of the high temperature elevation and several researchers have speculated that there should be a characteristic temperature (T_{out}) at which the outer structure is produced.[3–7] However, the mechanism and the characteristic temperature have not been elucidated until now. To control the effect of heat accumulation, it is important to elucidate the characteristic temperature as well as the temperature distribution. In this letter, we report that T_{out} and the temperature distribution during laser irradiation [$\Delta T(r)$] can be estimated from the morphology change of the heat modification by focusing femtosecond laser pulses with 250 kHz in glasses at various temperatures. By comparing the temperature dependence of the radius of the heat modification with the simulated temperature distribution, the temperature distribution and T_{out} were determined. From the obtained T_{out}, the mechanism of the modification due to heat accumulation was discussed.

We used amplified femtosecond laser pulses (250 kHz, 70 fs, 800 nm) of a mode-locked Ti-sapphire laser oscillator (Coherent; Mira and RegA). The femtosecond laser pulses were attenuated by a neutral density filter and focused inside a soda lime glass plate (Schott B 270 Superwite) through a 20× objective lens (NA=0.40; Nikon LU Plan ELWD 20×). The exposure time of laser irradiation (t_{ex}) was controlled by a mechanical shutter. The glass plate was placed on the stage in which temperature can be controlled by radiation from halogen lamps. The glass temperature was measured with a thermocouple.

Figures 1(a)–1(c) show the modifications inside a soda lime glass after 1.0 s irradiation by 250 kHz femtosecond laser pulses of 1.5 μJ/pulse at 23, 186, and 343 °C, respec-

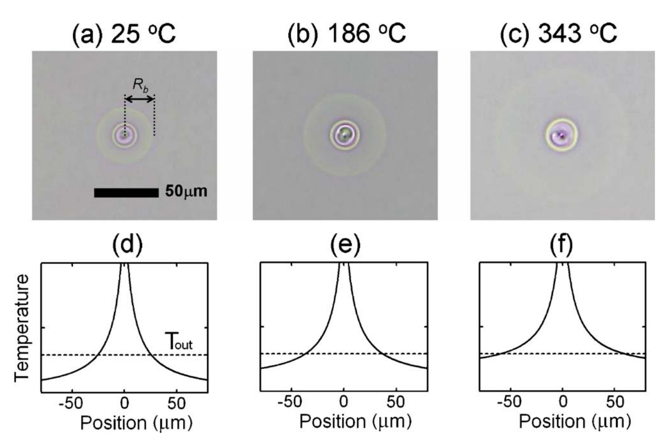

FIG. 1. (Color online) [(a)–(c)] Optical microscope images of the modification by 250 kHz femtosecond laser irradiation at various temperatures. [(d)–(f)]. The expected temperature distributions during laser irradiation for (a)–(c), respectively. The broken lines indicate the characteristic temperature (T_{out}) for the modification.

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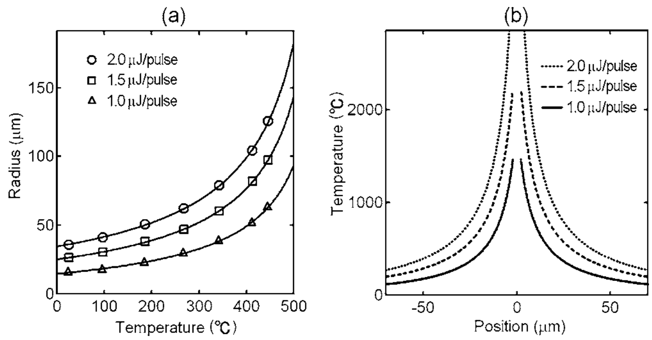

FIG. 2. (a) Relation between R_b and T_s at various pulse energies. The solid lines are fitting curves by Eq. (5). (b) Temperature distributions calculated by analyzing the relation between R_b and T_s by Eq. (4).

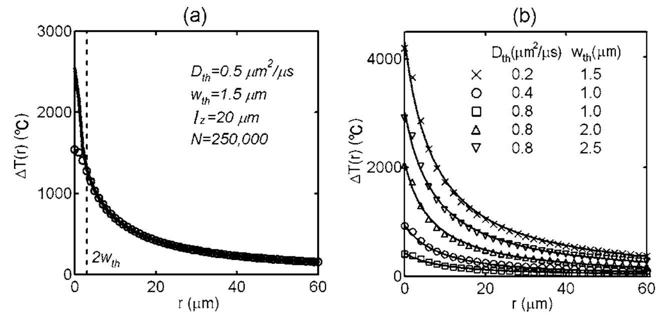

FIG. 3. (a) Calculated temperature distributions just before (open circles) and just after (solid line) irradiation of 250 000th femtosecond laser pulse. (b) Temperature distributions calculated with different D_{th} and w_{th}, and fitting curves by Eq. (3) (solid lines).

tively. As the temperature of glass (T_S) increased, the radius of the modification became larger. This temperature dependence supports the assumption that the volume of the modification should be determined by the characteristic temperature (i.e., T_{out}) because the temperature of the glass before irradiation enlarges the region in which the temperature exceeds T_{out} during laser irradiation [Figs. 1(d)–1(f)]. Figure 2(a) shows the plots of the radius (R_b) of the modified region at various pulse energies against glass temperature. The relation between R_b and T_S should reflect the temperature distribution during laser irradiation.

To obtain the temperature distribution from the relation between R_b and T_S, we searched for an equation that can express the temperature distribution during laser irradiation. First, the temperature distribution during laser irradiation at 250 kHz was simulated by a thermal diffusion model. The temperature distribution after N pulse irradiation under the assumption of constant thermal diffusion coefficient D_{th} can be written as

$$\Delta T(N\Delta t_L + t', r, z) = \sum_{n=0}^{N-1} \Delta T_1(n\Delta t + t', r, z), \tag{1}$$

where Δt_L is the time separation between laser pulses, t' is the time after the last irradiation, r is the radial distance from the beam axis, z is the position along the beam axis, and $\Delta T_1(t,r,z)$ is the temperature distribution change after irradiation of one femtosecond laser pulse. $\Delta T_1(t,r,z)$ was obtained by solving the thermal diffusion equation[14] under the initial temperature distribution of

$$\Delta T_0(r,z) = \Delta T_0 \exp\left[-\frac{r^2}{(w_{\mathrm{th}}/2)^2} - \frac{z^2}{(l_z/2)^2}\right], \tag{2}$$

where w_{th} and l_z are, respectively, the diameter and the longitudinal length of the initial heated volume and ΔT_0 is the maximum temperature. Because the modifications were observed from the laser propagation direction in Figs. 1(a)–1(c) and the radius of modification was defined by the maximum length from the beam axis, we evaluated the temperature distribution at $z=0$, where the temperature along the beam axis is maximum. Figure 3(a) shows the simulated temperature distributions just before and after the 250 000th irradiation. The distributions overlap at $r>2w_{\mathrm{th}}$ because the temperature change in this area is much smaller than that in the photoexcited region. This result means that the temperature sufficiently apart from the photoexcited region changes monotonically during laser irradiation. The temperature distributions after 250 000th and different w_{th} and D_{th} are shown in Fig. 3(b). Although the shapes of the distributions are different, all distributions at $r>2w_{\mathrm{th}}$ can be fitted by the same function,

$$f_{\Delta T}(r) = \frac{A}{(r-R_0)^2 + B}. \tag{3}$$

When the temperature of the glass before laser irradiation is T_S, the temperature distribution during laser irradiation can be written as

$$T(r,T_S) = f_{\Delta T}(r) + T_s = \frac{A}{(r-R_0)^2 + B} + T_S. \tag{4}$$

At the boundary of the modified region ($r=R_b$), the temperature is the same as the characteristic temperature T_{out} for creating the outer structure, i.e., $T(R_b,T_S)=T_{\mathrm{out}}$. Therefore, we can express R_b by A, B, R_0, T_S, and T_{out},

$$R_b(T_S) = R_0 + \left(\frac{A}{T_{\mathrm{out}} - T_S} - B\right)^{1/2}. \tag{5}$$

We fitted the relation between R_b and T_s by Eq. (5), and the fitting curves are shown in Fig. 2(a). From the fitting, we obtained $T_{\mathrm{out}}=560$ (± 20) °C. By substituting A, B, and R_0 into Eq. (4), the temperature distribution can be obtained. The temperature distributions are shown in Fig. 2(b).

To test the validity of the obtained $\Delta T(r)$ and T_{out}, we investigated the morphology change by simultaneous two laser irradiation at the spatially separated points inside the glass [Fig. 4(a)]. Because two heat sources exist in the simultaneous two laser irradiation, the morphology of the

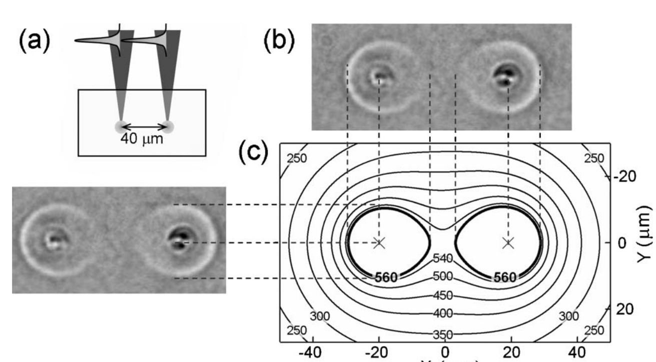

FIG. 4. Optical microscope images of the modification by two-beam irradiation and contour plot of the simulated temperature during laser irradiation.

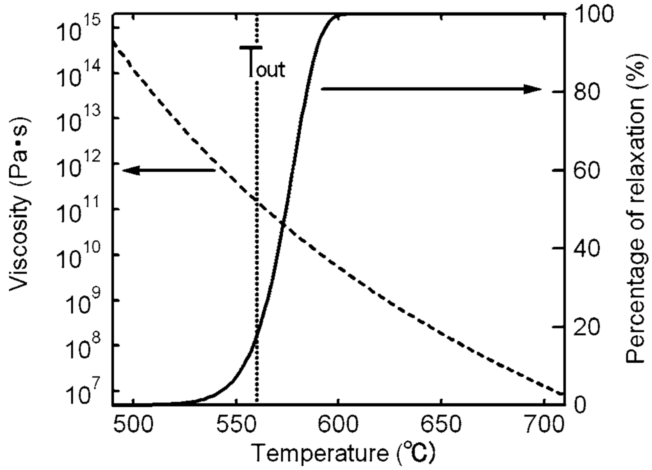

FIG. 5. Viscosity of B250 glass plotted as a function of temperature and percentage of relaxation during laser exposure time (=1 s). Fulcher's equation was used to interpolate the viscosity data of B250 from Schott.

modification should be mutually affected by the heat from the other irradiated point. Figure 4(b) shows the optical microscope image of the modification by the simultaneous irradiation of two 0.75 μJ femtosecond laser pulses in 1.0 s at 250 kHz inside a silicate glass. The tear drop shape of the modification indicates that a temperature gradient was created by the heat from the other irradiation point. To reproduce the modification in the simultaneous irradiation, we simulated the temperature distribution by the following equation:

$$T_{\mathrm{sim}}(r) = \Delta T(r) + \Delta T(r + 40\ \mu\mathrm{m}) + T_S, \tag{6}$$

which expresses that the materials at the 40 μm separated points are simultaneously photoexcited by femtosecond laser pulses. The simulated temperature distribution is shown in Fig. 4(c). The thick solid line, which corresponds to a contour line of $T = T_{\mathrm{out}}$ (560 °C), reproduces the tear drop shape of the modification. The consistency between the experiment and simulation in the case of two laser irradiation supports the validity of the obtained $\Delta T(r)$ and T_{out}.

The temperature distribution during laser irradiation and the T_{out} support our understanding of the mechanism of modification by highly repeated femtosecond laser irradiation. The physical meaning of the characteristic temperature $T_{\mathrm{out}} \sim 560$ °C we found for a silicate glass can be understood by the relaxation time of the glass after application of stress.[15] In glasses under high temperature, the relaxation after applying stress is considered by a viscoelastic model. According to the most simple viscoelastic model (Voigt–Kelvin element), the relaxation time is given by $\tau = \eta / G$, where η and G are viscosity and shear modulus, respectively. Because the temperature dependence of η of glass is much larger than that of G, we evaluated the relaxation of silicate glass during laser exposure time ($t_{\mathrm{ex}} = 1$ s) by temperature dependent η and a constant G(=29.3 GPa).[16] The viscosity and the calculated percentage of relaxation during laser irradiation ($P_{\mathrm{relax}} = 100\%[1 - \exp(-t_{\mathrm{ex}}/\tau)]$) plotted against temperature are shown in Fig. 5. We found that P_{relax} increases from ~1% to ~100% between 530 and 600 °C. T_{out} is almost in the middle of this temperature range. This means that the relaxation was almost complete at $r < R_b$, where the temperature exceeds T_{out} during laser irradiation, while the relaxation of the material in the outer region did not start.

Therefore, we propose the following mechanism of the structural change of glass during 250 kHz femtosecond laser pulse irradiation. (i) When femtosecond laser pulses are focused inside a glass at 250 kHz, thermal energy is accumulated in the photoexcited region. (ii) As a result of thermal diffusion, the materials surrounding the photoexcited region are heated, and the thermal expansion in the heated region gives the stress to the surrounding materials. (iii) The strain generates as relaxation in response to stress increase. (iv) The percentage of the relaxation depends on the distance from the photoexcited point because of the temperature distribution. In the region of $T < T_{\mathrm{out}}$, the laser exposure time is too short for the material to generate strain in response to stress increase. On the other hand, the relaxation almost finishes in the region of $T > T_{\mathrm{out}}$. (v) After the laser exposure is finished, the material is cooled quickly and the strain as the result of the relaxation remains in the region of $T > T_{\mathrm{out}}$.

In conclusion, we obtained the temperature distribution due to heat accumulation during 250 kHz femtosecond laser irradiation and the characteristic temperature of the heat modification. The obtained temperature distribution and characteristic temperature can explain the shape of the modification due to heat accumulation. According to the determined characteristic temperature, we found that the temperature dependence of the stress relaxation time is important to explain the mechanism of the structural change due to heat accumulation. The results in this study and the proposed mechanism will provide us with important information to control the structure by femtosecond laser pulses at a high repetition rate.

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